Abstract No. poul0509

Temperature dependent electron density distributions. The case of $Co_3(dpa)_4Cl_2$ (dpa = di(2-pyridyl)amide anion)

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Beamline(s): X3A1

Magnetism is one of the most important material properties in nature. Internationally, molecular magnetic materials have formed a very visible research area in recent years. Molecular magnetic materials, e.g. polynuclear transition metal complexes, can be described as single domaine magnets, with a domaine size in the nano regime. Therefore, they can be used to investigate magnetic phenomena on the nanoscale.

Magneto-chemical studies have shown that the $Co_3(dpa)_4Cl_2$ (dpa = di(2-pyridyl)amide anion) system has a drastic change in the magnetic moment as a function of temperature [1]. These changes have been correlated with changes in the metal-ligand bond lengths, and it has been postulated that the effect is due to spin cross over, i.e. a change in the metal orbital populations as a function of temperature. In order to obtain quantitative insight into the electronic structure of the system, and to gain a firmer understanding of its magnetic properties, we have measured multi-temperature single crystal X-ray diffraction data at beam line **X3A1** at **NSLS**. Data sets were collected at 20 K and room temperature at NSLS, whereas conventional data have been collected at 100, 150, 200 and 250 K on a Bruker SMART CCD diffractometer at Department of Chemistry, University of Aarhus.

We are currently modelling the electron density of the system (more than 100 unique atoms) with the multipole program XD. One problem has been to establish a good model for the three solvent molecules (CH_2Cl_2), which are present in the crystal structure. Once a reliable electron density model has been established we will extract experimental d-orbital population for the three Co centers based on the refined multipole populations. We will also carry out Bader topological analysis of the electron density, and e.g. use the topological properties to characterize the metal-metal bonds.

[1] Clerac et al, *J. Am. Chem. Soc*. **2000**, 122, 6226.

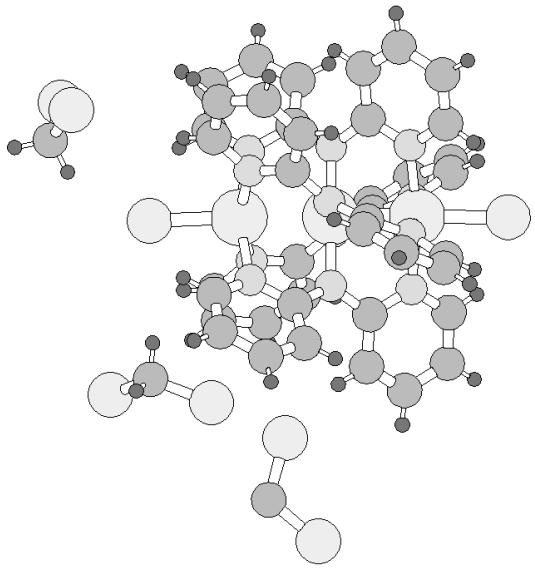

Figure 1. The molecular structure of $Co_3(dpa)_4Cl_2$.

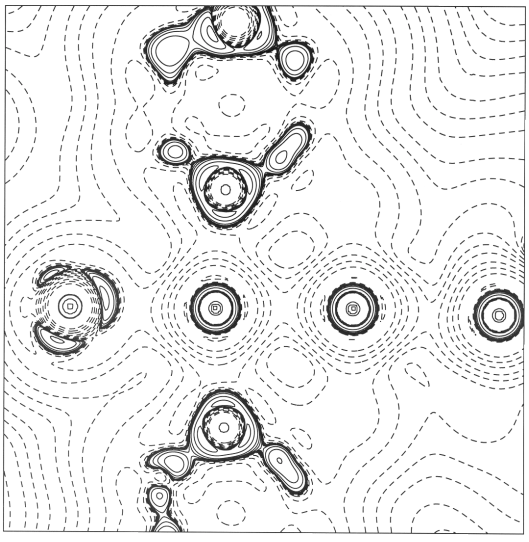

Figure 2. Negative Laplacian of the electron density